Hyperfine fields at the Ba site in the antiferromagnet $YBa_2Cu_3O_{6.05}$

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We report a Ba nuclear quadrupole resonance (NQR) study of the antiferromagnetic state of $YBa_2Cu_3O_{6.05}$ (Néel temperature $T_N = 415$ K) performed between 16 and 402 K. The Zeeman perturbed ^{137}Ba NQR spectrum yields information on two hyperfine fields present at the Ba site: the electric field gradient (EFG) and the internal magnetic field arising from the Cu(2) sublattice magnetization. The absolute value of the EFG is in remarkable agreement with cluster and band structure calculations thus demonstrating again that both methods provide a satisfying electronic bond picture for the Y-Ba-Cu-O compounds [except for the planar Cu(2) site]. The temperature dependence of the EFG arises from thermal expansion only. The internal field, $B(T)$, has been deduced from the modulation of the Ba spin-echo intensity. A calculation of the dipolar field at the Ba site produced by Cu(2) d electrons yields a value that is about three times larger than the experimental result. The discrepancy could be explained by assuming that part of the magnetic moment is located at oxygen ions. The temperature variation of $B(T)$ follows, up to 402 K, a power law $[B(0)-B(T)]/B(0)=AT^{\alpha}$ with $\alpha = 1.82(22)$ which agrees quite well with the result of a Cu(2) in-plane determination of the sublattice magnetization. Furthermore, this result is in accord with a spin-wave model for a quasi-two-dimensional (2D) antiferromagnet. The ''critical exponent'' β is estimated to be $\leqslant 0.18$ which is in accord with values proposed by models for 2D ordered magnetic systems. Thus $YBa_2Cu_3O_{6.05}$ behaves, in terms of its spin dynamics, as a quasi-2D antiferromagnet and this character can be studied either at out-of-plane Ba or at in-plane Cu(2) sites. [S0163-1829(96)06121-8]

I. INTRODUCTION

$YBa_2Cu_3O_6$ is the antiferromagnetic (AF) parent compound, with a Néel temperature, T_N, of about 415 K, of the $YBa_2Cu_3O_{6+x}$ superconductors (for $x>0.4$).[1] Since these materials are obtained by doping the AF structure an understanding of the magnetic properties of the latter is a prerequisite for the study of the mechanisms which cause superconductivity.

$YBa_2Cu_3O_6$ has a tetragonal crystal structure[2,3] and, according to neutron diffraction experiments, a three-dimensional (3D) AF structure[4,5] consisting of antiferromagnetically ordered Cu electronic spins in the CuO_2 plane [Cu(2) for short] that are themselves ordered antiferromagnetically along the tetragonal c axis. The magnetic moments arising from holes in the Cu(2) $3d_{x^2-y^2}$ orbitals, have a value of 0.64(3) μ_B/Cu(2) ion[6] and lie in the CuO_2 plane.[4,5,7,8] Although the magnetic structure is of 3D type, the AF coupling constants are strongly anisotropic: The superexchange interaction between nearest-neighbor spins in the CuO_2 plane is about five orders of magnitude stronger than the coupling *between* CuO_2 bilayers.[1] In other words, the ordered state exhibits a considerable quasi-two-dimensional (2D) character.

The degree of quasi-2D character of the AF state can be studied by nuclear magnetic resonance (NMR) and nuclear quadrupole resonance (NQR) techniques which allow one to measure the temperature dependence of internal hyperfine fields which are generally proportional to the sublattice magnetization. Indeed, such investigations have been performed in $YBa_2Cu_3O_6$ employing zero-field ^{63}Cu NMR (Refs. 9–11) and in La_2CuO_4 by studying the La site via zero-field ^{139}La NQR.[12,13]

In this paper, we demonstrate that the sublattice magnetization in $YBa_2Cu_3O_6$ can also be probed by measuring the internal magnetic field at the *out-of-plane* Ba site. The field is determined via the frequency of the spin-echo intensity modulation of pulsed ^{137}Ba NQR; preliminary results have been reported previously.[14] The technique allowed us to perform measurements up to 402 K, that is only 13 K below T_N, while previous Cu measurements reached 380 K. Our results support and complement previous[9–11] studies in $YBa_2Cu_3O_6$ and demonstrate that indeed this compound, together with La_2CuO_4, exhibits a strong quasi-2D character. This contrasts with our recent result[15] for the ''infinite-layer'' AF compound $Ca_{0.85}Sr_{0.15}CuO_2$ which has a stronger 3D character than $YBa_2Cu_3O_6$ and La_2CuO_4.

In addition to the sublattice study, we obtain information on the electric field gradient present at the Ba site. The results round off the general electronic bond picture for the Y-Ba-Cu-O structures.

This paper is organized as follows. The next section contains the necessary theoretical background of the procedure how to determine the internal field. Technical details including the preparation of the used sample are given in Sec. III. In Sec. IV, we present and analyze our data, followed by a discussion in Sec. V and a summary in Sec. VI.

II. THEORY OF THE EXPERIMENT

The Ba nuclear spins in $YBa_2Cu_3O_6$ interact with their electronic environment through electric and magnetic hyperfine couplings. In the presence of a magnetic field, for instance the internal magnetic field, $\mathbf{B}_{\rm int}$, due to the sublattice magnetization, the Hamiltonian of the Ba nuclear spin, $\mathbf{I}$, having a electric quadrupole moment, eQ, can be written as

$$\mathcal{H}=\mathcal{H}_{\text{Zeeman}}+\mathcal{H}_{\text{quadrupole}}, \tag{1}$$

with[16]

$$\mathcal{H}_{\text{Zeeman}}=-\gamma_n\hbar B_{\text{int}}(I_z\cos\theta+I_y\sin\theta\,\sin\phi+I_x\sin\theta\,\cos\phi) \tag{2}$$

and

$$\mathcal{H}_{\text{quadrupole}}=\frac{eQV_{zz}}{4I(2I-1)}\left[3I_z^2-I(I+1)+\frac{\eta}{2}(I_+^2+I_-^2)\right]. \tag{3}$$

Here, $V_{\alpha\alpha}$ ($\alpha=x,y,z$) denote the principal components of the electric field gradient (EFG) tensor, **V**, with the axes labeled according to the convention $|V_{xx}|\leqslant|V_{yy}|\leqslant|V_{zz}|$; x,y,z are chosen as the frame of reference. The asymmetry parameter of **V**, η, is defined as $\eta=(V_{xx}-V_{yy})/V_{zz}$, $\eta\in[0,1]$. In $YBa_2Cu_3O_6$, due to symmetry, one permutation of the x,y,z set coincides with the tetragonal **a**, **b**, and **c** crystal axes. θ and ϕ are the polar and azimuth angle, respectively, of $\mathbf{B}_{\text{int}}$ in this crystal frame. γ_n is the nuclear gyromagnetic ratio.

Due to the Ba tetragonal site symmetry, $\eta=0$ and V_{zz} is along the **c** axis. Since the Cu(2) magnetic moments lie in the CuO_2 plane, the symmetrical position of Ba with respect to the Cu(2) atoms requires the z component of $\mathbf{B}_{\text{int}}$ to be zero, that is $\theta=90°$. This is valid for each crystallite of our unoriented polycrystalline sample.

As we will show in Sec. IV, $\mathbf{B}_{\text{int}}$ is small, hence $\mathcal{H}_{\text{Zeeman}}\ll\mathcal{H}_{\text{quadrupole}}$, and Eq. (1) can be solved by first-order perturbation theory. For our special case of $I=3/2$, $\eta=0$, and $\theta=90°$, we obtain doubly degenerate $\pm\frac{3}{2}$ energy levels and a splitting of the $\pm\frac{1}{2}$ levels into the two levels $+\frac{1}{2}$ and $-\frac{1}{2}$. Transitions between these levels yield three resonance signals at frequencies

$$\nu_{\mp3/2\leftrightarrow\pm1/2}=\frac{eQV_{zz}}{2h}\pm\frac{\gamma_n}{2\pi}B_{\text{int}} \tag{4}$$

and

$$\nu_{-1/2\leftrightarrow+1/2}=\frac{\gamma_n}{\pi}B_{\text{int}}. \tag{5}$$

Here, $eQV_{zz}/2h$ is called the NQR frequency, ν_Q. For the ^{137}Ba isotope, the relevant numerical values are $eQ=0.28(3)\times10^{-28}$ m^2 (Ref. 17) and $\gamma_n=2.9728703(6)\times10^7$ rad (Ts)$^{-1}$.[18]

As we will see in Sec. IV, $\nu_{-1/2\leftrightarrow+1/2}$ is very small and the two frequencies $\nu_{-3/2\leftrightarrow+1/2}$ and $\nu_{+3/2\leftrightarrow-1/2}$ overlap, thus not allowing to measure B_{int}. Instead, we have determined B_{int} by utilizing the effect that the presence of a small static magnetic field (either internal or external) leads to a *modulation* of the spin-echo intensity.[19,20] The spin echo is obtained by a conventional pulse experiment with an exciting pulse followed, at time τ, by a refocusing pulse; the spin-echo maximum then appears at time 2τ. This is the same method as employed to determine the spin-spin relaxation time, T_2. In the case of $\theta=90°$, $\eta=0$ and $I=3/2$, the intensity of the spin echo at time 2τ is given by the relation[21,22]

$$I(2\tau)=I_0\exp\left(-\frac{2\tau}{T_2}\right)[1+\cos(2\tau\omega_{\text{int}})]. \tag{6}$$

The term $I_0\exp(-2\tau/T_2)$ describes the general spin-echo decay due to spin-spin relaxation processes while the $\cos(2\tau\omega_{\text{int}})$ term produces the modulation with angular frequency

$$\omega_{\text{int}}=\gamma_nB_{\text{int}}. \tag{7}$$

It turned out that in case of the Ba NQR in $YBa_2Cu_3O_6$, Eq. (6) had to be replaced by the following relation:

$$I(2\tau)=I_0\exp\left(-\frac{2\tau}{T_2}\right)\exp\left(-\frac{2\tau}{T_2'}\right)\times\left[1-K\,\exp\left(-\frac{2\tau}{T_2''}\right)\cos(2\tau\omega_{\text{int}})\right]. \tag{8}$$

The factor $\exp(-2\tau/T_2')$, already introduced in Ref. 22, takes into account, via an effective time constant T_2', the distribution of dipolar fields and EFG's (due to an inherent disorder of the structure) at the various Ba sites. The factor $\exp(-2\tau/T_2'')$, used in a similar fashion in Ref. 23, reflects the fact that the amplitude of the modulation decreases the faster the larger the asymmetry factor, η. In other words, T_2'' arises from a distribution of η values because of structural disorder in the Cu(1) chains due to residual oxygen which was not removed during the deoxidation process (see Sec. III). Finally, the constant K must be introduced since the broad NQR spectrum, consisting of an overlap of the two $\mp\frac{3}{2}\leftrightarrow\pm\frac{1}{2}$ resonance signals, allows only a fraction of all Ba spins to participate in the modulation effect. We will come back to this point in Sec. IV. However, it should be stressed already here that these various factors do not affect the value of ω_{int}.

III. EXPERIMENTAL DETAILS

The ^{137}Ba NQR experiments were performed in *zero external* magnetic field using a home-built standard phase-coherent pulsed NMR spectrometer with 1 kW output power. In all our resonance experiments, we used a resonance circuit with a rather small coil (6 mm diameter, 15 mm length) to achieve, if necessary, very short optimal pulses and thus broad spectral excitation. To prevent sparking, we kept the probe head in nitrogen or helium gas at a pressure of 3 bar.

The resonance signals were obtained by a phase alternating add-subtract spin-echo[24] technique, similar to that used in Ref. 14 to eliminate the ringing after the pulses and to improve the signal-to-noise ratio. By scanning the spectrometer frequency and retuning the resonance circuit in 50 kHz steps, the spin-echo signals were measured in quadrature and then integrated over time (called spin-echo intensity) yielding in a point-wise fashion the rather broad ^{137}Ba NQR spectra as shown in the inset of Fig. 1.

When determining the ^{137}Ba NQR frequency which corresponds to the center of the spectrum, we used pulses with optimal length, 3.5–5.5 μsec, to maximize the spin-echo intensity. When measuring the modulation frequency, we used shorter pulses (1.2–2.5 μsec) in order to excite the largest

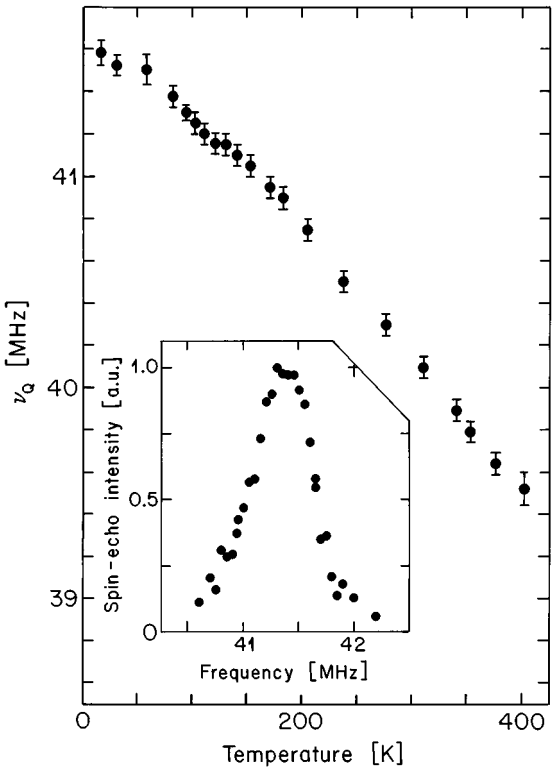

FIG. 1. Temperature dependence of the ^{137}Ba NQR frequency, ν_Q, in $YBa_2Cu_3O_{6.05}$. Inset: NQR spectrum at 81.5 K.

possible part of the Ba signal and to maximize the amplitude of the spin-echo modulation.

Any external magnetic field, in particular the earth's magnetic field, would cause an additional modulation of the spin-echo intensity as observed, for instance, in our T_2 measurements of Ba in the superconductor $YBa_2Cu_4O_8$.[25] To avoid such artifacts, we have screened the cryostat (which contains the NQR probe head) with a 1 mm thick μ metal forming a half-open cylinder. Thus the external magnetic field at the sample site could be reduced to about 5×10^{-7} T as measured by a Hall probe. The sample temperature was stabilized and measured with a precision of $\pm$ 0.5 K.

Our experiments were carried out with an unoriented large-grain size $YBa_2Cu_3O_{6.05}$ powder sample, which was prepared by deoxidation of $YBa_2Cu_3O_7$ material for 18 h at 780 °C and at a pressure of 0.02 mbar. The oxygen content of 6.05(5) per unit formula was determined by thermal gravimetry and by x-ray powder diffraction. Our values for the lattice constants, namely $a=b=3.856(4)$ Å and $c=11.813(10)$ Å, agree with literature data.[26,27]

The starting $YBa_2Cu_3O_7$ material had been synthesized by the standard solid state reaction.[28] $BaCO_3$ with 90% enriched ^{137}Ba had been used to improve the NQR signal intensity since the natural abundance of ^{137}Ba is only 11.3%.

IV. RESULTS AND ANALYSIS

As mentioned above, the two Zeeman perturbed NQR ^{137}Ba signals with frequencies $\nu_{+3/2\leftrightarrow-1/2}$ and $\nu_{-3/2\leftrightarrow+1/2}$ [Eq. (4)] could not be resolved. Instead, we observed a ''single line'' spectrum as shown in the inset of Fig. 1.

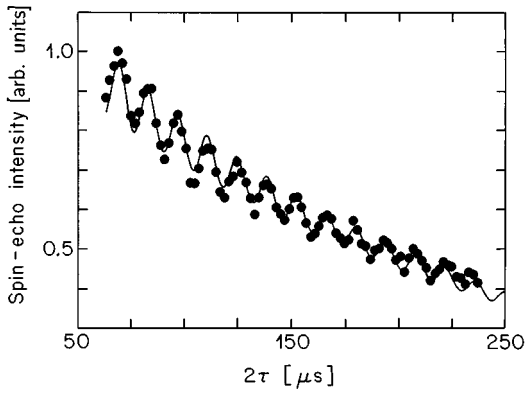

FIG. 2. ^{137}Ba spin-echo intensity in $YBa_2Cu_3O_{6.05}$ at 58 K as a function of 2τ (see text). The solid line is a fit of Eq. (8) to the data.

A. The NQR frequency

According to Eq. (4), the Ba NQR frequency, ν_Q, is given by the mean value of $\nu_{+3/2\leftrightarrow-1/2}$ and $\nu_{-3/2\leftrightarrow+1/2}$. Therefore, ν_Q corresponds to the center of the ''single line'' spectrum. At 16 K, the lowest temperature we used, the NQR frequency is 41.58(6) MHz and then decreases with rising temperature to 39.53(8) MHz at 402 K as shown in Fig. 1.

B. The internal field at the Ba site

A typical example of the modulation of the spin-echo intensity as a function of 2τ is represented in Fig. 2. The solid line is a fit of Eq. (8) to the data thus yielding, besides the various time constants, the modulation frequency, $\omega_{\rm int}$, and hence the internal magnetic field, B (where we have dropped the index ''int'').

At 16 K, B is $(1.54\pm0.02)\times10^{-2}$ T; it then decreases monotonically with rising temperature to $(8.1\pm0.9)\times10^{-3}$ T at 402 K. The temperature dependence of B is shown in Fig. 3. The solid line is a fit to be discussed later.

As mentioned before, we have chosen the particular function for the spin-echo decay, $I(2\tau)$, as given by Eq. (8), because of various physical arguments that explain the decay rate and the relatively small modulation amplitude. Our main interest, however, concerns the modulation frequency, $\omega_{\rm int}$. And this value is not critically affected by details of the form of $I(2\tau)$. For instance, instead of using Eq. (8), the data may be fitted with a function like $I(2\tau)\propto\exp(-2\tau/T_2''')[1+K'\cos(2\tau\omega_{\rm int})]$. Although this fit is not as perfect as that provided by Eq. (8) and the reproduction of the decay of the spin-echo intensity modulation is not satisfying, it yields the same $\omega_{\rm int}$, the difference being less than 1%.

A second important point is that the *value* of $\omega_{\rm int}$ is independent of the pulse length (in the range of pulse lengths we have used) although choosing the proper pulse length is essential for the modulation to be observed at all.

V. DISCUSSION

We will discuss the value and the temperature dependence of both the NQR frequency and the internal magnetic field.

A. The NQR frequency

At present, there are two quite successful theoretical approaches that allow one to calculate the NQR frequency in

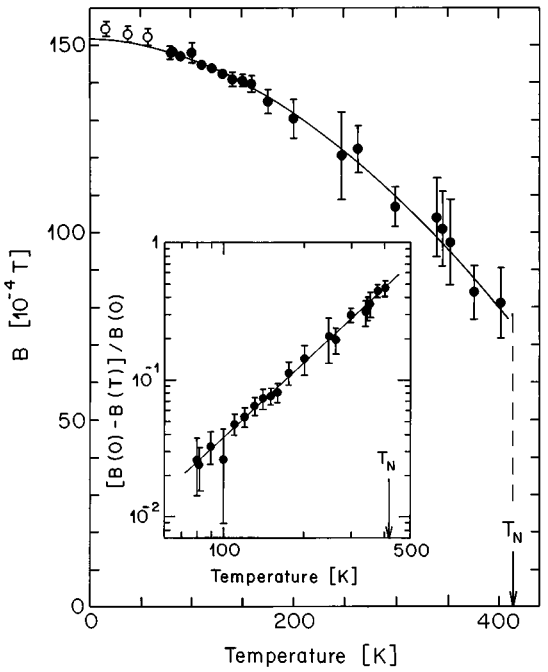

FIG. 3. Temperature dependence of the internal magnetic field, B, at the Ba site in $YBa_2Cu_3O_{6.05}$. The three data points denoted by open circles were not included in the fit (see text). T_N denotes the Néel temperature. Inset: Fit to the log-log plot of $[B(0)-B(T)]/B(0)$ vs T.

solids such as the high-temperature superconductors and their AF parent compounds: the Hartree-Fock cluster method and the band structure calculation using the ''full potential linearized augmented plane waves'' (LAPW). The first method predicts for $YBa_2Cu_3O_6$, at room temperature, a ^{137}Ba NQR frequency of 39.8 MHz,[29] and the second, at 373 K, 37.2 MHz.[30] Both results, in particular the first one, are in good agreement with the experimental data, for instance with the value of 40.10(5) MHz for 310 K. This agreement underlines once again that both methods provide a satisfying electronic bond picture for the compounds involved, except for the well-known discrepancy of the planar Cu(2) data. The good agreement is probably not surprising in view of the ''pure ionic'' character of the Ba bonding in the $YBa_2Cu_3O_6$ structure.

Since temperature dependent cluster and band structure calculations of NQR frequencies do not exist for $YBa_2Cu_3O_6$, we will use the simple point charge model to explain the decrease of ν_Q with rising temperature. In this semiempirical approach, one assumes that the EFG tensor can be written as the sum of two terms, a lattice and a valence contribution. The lattice contribution arises from all charges outside the ion under consideration and the valence contribution from nonfilled shells of the subject ion. Due to the closed electron shell of the Ba^{2+} ion, ν_Q of Ba is exclusively determined by the lattice contribution to the EFG and is as such very sensitive to structural changes.

Using the lattice parameters given in the literature[2,3] and taking integer values for the valence of the different ions we calculated a reduction of ν_Q of about 6% for the temperature range 10 to 373 K. This result is in good agreement with the observed decrease of $\nu_Q(T)$. We thus conclude that the temperature variation of ν_Q arises from thermal expansion only.

B. The internal field at the Ba site

As our experiments have shown there is an internal magnetic field present at the Ba site. This result is at odds with two other studies. In the first one, it had been erroneously stated[31] that due to symmetry the internal fields at the Ba site must cancel and that the ''narrow'' ^{135}Ba NQR signal the authors observed would support this argument, although the width of the signal is nearly 1 MHz. Apparently, it had been overlooked that the signal could be and indeed is, as we have shown for the ^{137}Ba isotope, an overlap of two signals.

In the second study, time-differential perturbed angular correlation (TDPAC) measurements[32] are reported and the magnitude of the field at the Ba site is quoted to be about 1.2 T, which is two orders of magnitude larger than our value. If the internal field would be this large, the signals with frequencies $\nu_{-3/2\leftrightarrow+1/2}$ and $\nu_{+3/2\leftrightarrow-1/2}$ would have been well separated; however, we only observed a single spectrum. Furthermore, the intensity modulation of our signal would not have occurred if the signal would be only *one* of the two $\nu_{-3/2\leftrightarrow+1/2}$ and $\nu_{+3/2\leftrightarrow-1/2}$ lines, since both lines would not have been excited simultaneously.

When checking Ref. 32 for possible reasons for the tremendous discrepancy between our value for the internal field and the TDPAC result, one notes the following: (i) The authors comment neither on possible radiation damages when implanting 30 keV Ba ions, nor on the ions' penetration depth, which should be large compared to the unit cell dimensions, and they do not provide evidence that the Ba ions occupy the regular Ba sites. (ii) Only TDPAC data points were given without drawing the fit function that is supposed to yield the 1.2 T value. In addition, data cover only a small time window, which makes identification of the oscillations rather difficult. (iii) The authors admit that it is ''difficult to estimate the interaction frequency'' (which yields the internal field). We therefore conclude, that the result of the TDPAC study is doubtful.

We will now discuss calculations of the internal field at the Ba site. In a first approach, we assumed point like magnetic moments of 0.64(3) μ_B (Ref. 6) at the Cu(2) sites. Summing over the four next Cu(2) neighbors yields a dipolar field of $(6.25\pm0.29)\times10^{-2}$ T pointing perpendicular to the c axis. This value is about four times larger than the experimental result obtained at 16 K. Extending the summation to 16 AF Cu atoms in the nearest-neighbor CuO_2 plane and four atoms in the next-nearest plane, yields 4.7×10^{-2} T (Ref. 33), still a factor of 3 too large. Extending the summation even further would give a result that oscillates around a value somewhere above 4.7×10^{-2} T.

In a second approach,[33] extended $3d_{x^2-y^2}$ electrons are assumed and their dipolar field is computed by evaluating the integrals over the spherical functions of these electrons. However, the effect of finite size and anisotropy of the Cu d electrons is only in the order of a few percent.[33] A possible explanation of the discrepancy between these values and the experimental result could be the copper-oxygen covalency. If one assumes that part of the magnetic moment is located at

oxygen ions, which corresponds to an extension of the ''magnetic moment distribution,'' the dipolar field at the Ba site would be reduced.

We now turn to the temperature dependence of the internal field. The usual procedure is to fit such data by a power law,

$$\frac{[B(0)-B(T)]}{B(0)}=AT^{\alpha}, \tag{9}$$

where $B(0)$ denotes the field at 0 K.

Obviously, this procedure is only applicable if no ''anomalies'' like phase transitions are present in the temperature range studied. A close inspection of our data in Fig. 3 reveals that some sort of anomaly occurs between 60 and 80 K although the origin of the slight kink in the data is still unknown. However, above this temperature up to 402 K, the data behave in a smooth way and we have fitted them by Eq. (9). The fit as represented by the solid line in Fig. 3 yields $\alpha=1.82(22)$, $A=(8.7\pm2.0)\times10^{-6}$ $K^{-\alpha}$, and $B(0)=(1.52\pm0.02)\times10^{-2}$ T. The inset in Fig. 3 shows a fit to the log-log plot of $[B(0)-B(T)]/B(0)$ vs T.

A power law behavior of the internal field had already been found for the in-plane Cu(2) site in $YBa_2Cu_3O_6$ as measured by zero-field ^{63}Cu NMR with α values of 1.87,[9] 1.93 (Ref. 10), and 1.94.[11] Our value of α for the out-of-plane Ba site in $YBa_2Cu_3O_6$ is in good agreement with those values; thus the internal field probed either at in-plane or out-of-plane sites exhibits the same temperature dependence, i.e., the same exponent α.

The internal field is related to the sublattice magnetization, $M(T)$, by the equation $M(T)/M(0)=B(T)/B(0)$ if we assume the hyperfine coupling constants to be independent of temperature. Matsumura *et al.*[10] have calculated the quantity $[M(0)-M(T)]/M(0)$ in the framework of a spin-wave model, with four spin wave modes, for a quasi-2D antiferromagnet using exchange coupling parameters reported by neutron scattering experiments. The authors calculated *numerically* $[M(0)-M(T)]/M(0)$ which yields, above 25 K and up to 200 K, a power law with $\alpha=1.93$.

The agreement between model and experiment is taken as strong evidence that, in terms of spin dynamics, $YBa_2Cu_3O_6$ behaves as a quasi-2D antiferromagnet. The good agreement between Matsumura's $\alpha=1.93$ and our value of 1.82(22) for the temperature dependence as probed at the *out-of-plane* Ba site demonstrates that the quasi-2D AF character of $YBa_2Cu_3O_6$ can also be studied at an out-of-plane site yielding the same value for α.

An out-of-plane study of the internal field in the AF compound La_2CuO_4 has recently been reported.[13] By zero-field ^{139}La NQR, i.e., similar to our Ba NQR, the internal field had been measured yielding a temperature dependence which also obeys a power law with α of about 1.9 below 190 K. Thus this result fits into the general picture that $YBa_2Cu_3O_6$ and La_2CuO_4 exhibit a quasi-2D AF character. Because of experimental difficulties, one is still waiting for an in-plane measurement in La_2CuO_4 via Cu nuclei.

The 2D AF character of $YBa_2Cu_3O_6$ should also show up in the critical behavior of $M(T)$ near T_N where $M(T)$ is expected to obey the ''critical equation'' $M(T)\propto(1-T/T_N)^{\beta}$. So far, we succeeded to measure the internal field at the Ba site only up to 13 K below T_N, although this is the closest approach to T_N made so far by NMR-NQR. Obviously, the long range AF order and hence the internal field must disappear above T_N. If we assume that $B(T)$ decreases to zero in the range 402 K to T_N rather than performing a jump to zero at T_N, we estimate the critical exponent β to be equal or smaller than 0.18. The case *smaller* would apply if $B(T)$ would continue to obey Eq. (9) for a few more degrees beyond 402 K.

Models for ordered magnetic systems[34] predict β values of 0.325(2), 0.346(2), and 0.365(3) in case of the 3D Ising, XY, and Heisenberg model, respectively, while a 2D Ising model requires $\beta=0.125$. Obviously, our estimate of $\beta\leqslant0.18$ comes closer to 2D than to 3D values and thus supports our conclusion drawn from the general temperature behavior of the internal field. It should be noted that a similar situation applies to La_2CuO_4 where β is estimated to be $\lesssim0.1$ (Ref. 12) and $\lesssim0.15$.[13]

VI. SUMMARY

We have measured, as a function of temperature, the Zeeman perturbed ^{137}Ba NQR spectrum in $YBa_2Cu_3O_{6.05}$. The center frequency of the signal yields the NQR frequency, ν_Q, and hence the electric field gradient at the Ba site. The temperature dependence of ν_Q arises from thermal expansion only. The absolute value of ν_Q, for instance at room temperature, is in remarkable agreement with cluster and band structure calculations thus demonstrating again that both methods provide a satisfying electronic bond picture for the Y-Ba-Cu-O compounds [except for the planar Cu(2) site].

The spin-echo intensity modulation of the ^{137}Ba NQR signal allows to determine the internal magnetic field, $B(T)$, at the Ba site arising from the Cu(2) sublattice magnetization. Calculating the dipolar field at the Ba site produced by Cu(2) d electrons yields a value that is about three times larger than the experimental result. The discrepancy could be explained by assuming that part of the magnetic moment is located at oxygen ions.

The temperature variation of $B(T)$ follows, up to 402 K, a power law $[B(0)-B(T)]/B(0)=AT^{\alpha}$ with $\alpha=1.82(22)$ which agrees quite well with the exponent extracted from the Cu(2) in-plane determination of the sublattice magnetization. Furthermore, this result is in accord with a spin-wave model for a quasi-2D antiferromagnet with strongly anisotropic exchange coupling constants.[10] Thus $YBa_2Cu_3O_{6.05}$ behaves, in terms of its spin dynamics, as a quasi-2D antiferromagnet and this character can be studied either at out-of-plane Ba or at in-plane Cu(2) sites.

This conclusion is also supported by an estimate of the critical exponent β assuming that $B(T)$ obeys, near T_N, the ''critical equation'' $B(T)\propto(1-T/T_N)^{\beta}$. The result, $\beta\leqslant0.18$, is in accord with values proposed by models for 2D ordered magnetic systems, e.g., the 2D Ising model with $\beta=0.125$.

In conclusion, our results for $YBa_2Cu_3O_{6.05}$ support and complement the picture that this compound as well as La_2CuO_4 can be regarded as quasi-2D antiferromagnets. This contrasts with our recent findings[15] for the ''infinite-layer'' structure $Ca_{0.85}Sr_{0.15}CuO_2$ which is more appropriately described as a 3D antiferromagnet.

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